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Enhanced spin accumulation in Fe$_3$O$_4$ based spin injection devices below the Verwey transition

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Abstract
Spin injection into GaAs and Si (both n and p-type) semiconductors using Fe$_3$O$_4$ is achieved with and without a tunnel barrier (MgO) via three-terminal electrical Hanle measurement. Interestingly, the magnitude of spin accumulation voltage ($\Delta V$) in semiconductor is found to be associated with a drastic increment in $\Delta V$ in Fe$_3$O$_4$ based devices for temperature < 120 K (T$_V$, the Verwey transition). Such an enhancement of $\Delta V$ is absent in the devices with Fe as spin source. Further, the overall device resistance has no drastic difference at T$_V$. This renders a direct proof that the observed $\Delta V$ is not influenced by the so-called metal-to-insulator transition of Fe$_3$O$_4$ at T$_V$. Observations from our elaborate investigations show that spin polarization of Fe$_3$O$_4$ has an explicit influence on the enhanced spin injection. It is argued that the theoretical prediction of half-metallicity of Fe$_3$O$_4$ above and below T$_V$ has to be reinvestigated.

Introduction
Electrical spin injection and detection using three-terminal Hanle (3TH) technique in semiconductors have attracted a lot of attention in the past few years owing to the successful demonstration of spin injection at room temperature in various systems [1–6]. In the field of semiconductor spintronics devices, spin injection and detection in various materials find a direct application, for example the proposed Datta-Das transistor [7]. It is important to have high spin injection and detection efficiency in order to obtain a good signal-to-noise ratio in any device. Many groups have adopted different techniques like use of tunnel barriers [1–6] and/or materials with high spin polarization such as Heusler alloys [8] and Fe$_3$O$_4$ [9], in an effort to enhance the electrical spin injection efficiency at room temperature in various systems.

Magnitude of spin accumulation in any system depends on factors like (i) type of the injector (ii) spin polarization of the ferromagnet/insulator combination (iii) junction resistance (iv) the uniformity of film at the interface, etc. Choosing a suitable spin injector for an efficient spin injection is one of the essential criteria. Materials like Fe$_3$O$_4$ and La$_{2/3}$Sr$_{1/3}$MnO$_3$ are promising owing to their high spin polarization when compared to their counterparts, ferromagnetic metals such as Fe, Co, CoFe etc. However, though theoretical calculations indicate half-metallicity in Fe$_3$O$_4$ crystal [10, 11], spectroscopic experimental observations always show a spin polarization ranging from 40% to 80% at room temperature depending on the orientation, surface morphology and methods of preparation of Fe$_3$O$_4$ [12–14]. Similarly, indirect probing of temperature dependent spin polarization from magnetoresistance measurements reveal maximum spin polarization at Verwey transition (T$_V$ ~ 120 K) [15, 16]. Hence, the theoretical prediction of half-metallicity of Fe$_3$O$_4$ thin film at room temperature is still under debate.

As the name indicates, 3TH devices consist of 3 terminals for the application of current and the measurement of voltage with one common lead at the center which is the device. Schematic of a typical 3TH measurement for electrical spin injection and detection is as shown in figure 1(a). The magnetic layer at the top (i.e. Fe$_3$O$_4$ in this case) acts as a spin source which is magnetized to the saturation magnetization in the plane of the layer. Application of a constant current through the Fe$_3$O$_4$ thin film makes sure that sufficient spin-polarized carriers are pumped into the semiconductor where the decay of spin polarized carriers will be measured.
Insulators are generally used in between metal/semiconductor junctions for better electrical spin injection and detection. In the absence of these tunnel barriers, devices are known to suffer from a conductivity mismatch problem \[17, 18\]. However, even Schottky barriers are known to be having similar behavior like tunnel barrier \[19–21\]. Once, the spins are injected into the semiconductor, these accumulated spin-polarized carriers at the junction of semiconductor get scattered or precess depending on the type of semiconductor. To be precise, for example, doping of semiconductor results in the momentum scattering and thus the scattering of spins in the lattice. Whereas, the internal magnetic field due to the spin–orbit coupling from the material results in the precessional motion of spins and thus leads to the spin scattering. Thus, there is a gradient in the measured voltage between the device and the far end of semiconductor due to the spin scattering process within the semiconductor. The accumulated voltage right at the interface of device region with respect to the ground (\(\Delta V\)) depends on how fast the spin-polarized carriers get relaxed (i.e. spin relaxation time) and how far these carriers can traverse within the semiconductor (i.e. spin diffusion length). In order to scatter the spins in a controlled manner inside the semiconductor to have a better control over the rate of spin relaxation, a perpendicular magnetic field \(B\) to the device plane is applied. As the strength of \(B\) is increased, spins are forcefully dephased from the initial direction of in-plane, and thus resulting in reduced \(\Delta V\). The spin accumulation voltage signal is found to decay as a Lorentzian function of \(B\), \(\Delta V(B) = \Delta V(0)/(1 + (\omega_L \tau)^2)\), where \(\omega_L = g\mu_B B/\hbar\), is the Larmor frequency of spin precession and \(\tau\) as effective spin relaxation time. Decay of spin signal \(\Delta V\) in the semiconductor with perpendicular magnetic field is depicted in figure 1(b). Decay of spin signal \(\Delta V\) in the semiconductor with perpendicular magnetic field is depicted in figure 1(b). If we look at the magnitude of the accumulation voltage (\(\Delta V\)) in 3TH geometry of measurement carefully, then \(\Delta V\) is governed by the factor, \(\Delta V = \text{TSP} \times \frac{\Delta \mu}{2}\), where TSP is the tunnel spin polarization of the ferromagnet/insulator combination and \(\Delta \mu\) the difference in chemical potential for up-spins and down-spins, which is nothing but the spin accumulation at the injected material \[1\]. Hence, in this regard, spin polarization of the magnetic material together with barrier material (if used) determines the spin accumulation at the interface of the semiconductor.

Based on these discussions, we have demonstrated the electrical injection and detection of spins at room temperature in semiconductors like GaAs and Si using \(\text{Fe}_3\text{O}_4\) with the help of 3TH technique \[9, 22\].

**Methods**

In this report, the study of the spin accumulation voltage as a function of temperature is carried out from the experiments based on electrical spin injection and detection. The devices of interest are ranging from an epitaxial injection system \(\text{Fe}_3\text{O}_4\)/MgO/GaAs to polycrystalline \(\text{Fe}_3\text{O}_4\)/MgO/Si junctions with tunnel barriers, including
the textured Fe3O4/GaAs Schottky junctions. Pulsed laser ablation technique is used to prepare the thin film devices of Fe3O4 on GaAs and Si substrates which are deposited under identical temperature of growth (450 °C) and pressure (base vacuum \( \sim 10^{-5} \) mbar) with the MgO deposition temperature being 500 °C. Subsequently, device structures are fabricated using photo-lithography with structure details similar to the dimensions discussed in [9]. As mentioned earlier, in any 3-terminal device, the spin accumulation voltage is measured as a function of perpendicular magnetic field \( B \) at different temperatures. A typical temperature dependent \( \Delta V \) versus \( B \) of Fe3O4/MgO/GaAs (n-type \( 10^{18} \text{ cm}^{-3} \)) device is shown in figure 1(c). As the temperature decreases, the FWHM of the spin signal is reduced due to the lower scattering rate of spins in GaAs. The Lorentzian fit (solid lines in figure 1(c)) to temperature dependent spin signal, \( \Delta V \), can be used to extract the spin relaxation time in GaAs depending on the type of doping at a given temperature [9]. An extensive study of the temperature dependence of spin injection and detection in different devices of Fe3O4/MgO/Si (both n-type \( 1 \times 10^{19} \text{ cm}^{-3} \) and p-type \( 3 \times 10^{18} \text{ cm}^{-3} \)) and Fe3O4/GaAs (n-type \( 10^{18} \text{ cm}^{-3} \)) Schottky systems show a systematic variation of spin-relaxation time for different doping concentrations of substrates [22]. Even in the absence tunnel barrier material, spin injection and detection is shown to be equally possible, as is evident from the earlier studies [21]. However, the interesting observation of spin injection voltage (\( \Delta V \)) as a function of temperature allowed us to compare various systems at different temperatures. Strikingly, we have found that there is an anomalous increment of \( \Delta V \) below Verwey transition. Control experiment using Fe/MgO/GaAs devices shows us that the increase in \( \Delta V \) is unique to Fe3O4 based spin injector devices.

**Results and discussions**

We have investigated the spin accumulation in our devices, like (i) (100) oriented Fe3O4/MgO/n-type GaAs, (ii) polycrystalline Fe3O4/MgO/n-type Si, (iii) (111) oriented Fe3O4/n-type GaAs and (iv) (100) oriented Fe/MgO/n-type GaAs systems (either using metallic or oxide magnetic material as a spin injector into systems like GaAs and Si with and without MgO as a tunnel barrier) [9, 22]. In this report, the magnitude of spin accumulation voltage \( \Delta V \) in both GaAs as well as Si is studied as a function of temperature with Fe3O4 as the spin injector. These observations are then compared with the 3TH voltage obtained from the devices where Fe is used as a spin source. Plot of \( \Delta V \) as a function of temperature for different injection systems (having same device area of \( 100 \times 200 \mu m^2 \)) with a constant injection current of \( 1 \mu A \) for GaAs and \( 100 \mu A \) for a device area of \( 100 \times 200 \mu m^2 \) are given in figure 2.

There is a sharp increase in the spin accumulation at \( T < 120 \) K for the devices with Fe3O4 as a spin injector, which coincides with \( T_V \) of Fe3O4, \( T_V \sim 122 \) K [23] is a structural transition in Fe3O4 lattice, which changes the magnetization, resistivity, heat transport etc drastically. As seen from figure 2(d), device with Fe as a spin injector has no such type of enhancement of \( \Delta V \) at around 120 K which otherwise exists in Fe3O4 based devices (figures 2(a)–(d)). Normalized spin accumulation voltage is plotted in figure 2(e) for different junctions at various temperatures and the comparison of the magnetization versus temperature is shown in figure 2(f). Shaded area in the figure is the region of the Verwey transition of Fe3O4 in different films used, which coincides with the region of enhancement of \( \Delta V \). In Fe3O4 spin injection system, the increase in the value of \( \Delta V \) at \( T_V \) was found to be between 3.3 to 4 times higher irrespective of the relaxation media (GaAs or Si), type of barrier (tunnel or Schottky barriers), the orientation; (100), (111) and crystallinity of Fe3O4 used. Generally, a gradual enhancement of spin accumulation is observed with the increase of barrier resistance in the case of metallic spin injector systems [6, 24, 25]. Even the enhancement in the spin signal from room temperature to 5 K observed for a given barrier thickness is feeble in different cases [2–4, 6, 21, 26].

The magnitude of spin accumulation voltage \( \Delta V \) differs in various injection systems (figures 2(a)–(d)) though the injection current and device area were kept constant for all GaAs devices. This is purely because of the variation of conditions across the film stack for different devices. In our case, the observed electrical spin accumulation voltage is higher in Fe3O4/GaAs junctions (2.2 mV for \( 1 \mu A \) at 300 K), whereas it is least in the case of Fe3O4/MgO/Si (10.7 mV for \( 100 \mu A \) at 300 K). \( \Delta V \) is found to be 0.024 mV at 300 K for Fe/MgO/GaAs device, which is comparatively lower by 2 orders of magnitude when compared with Fe3O4/GaAs junctions and 1 order lower than Fe3O4/MgO/GaAs devices (0.31 mV for \( 1 \mu A \) at 300 K). Hence, the spin accumulation at various device interfaces is found to be different which is purely dependent on the interface conditions [22].

In our case, resistance of the spin injector Fe3O4 is highly temperature dependent and the presence of Schottky or the tunnel barrier always ensures the resistance on either side of the barrier to be irrelevant even at temperatures lower than \( T_V \) of Fe3O4. Here, we point out that though the resistance change in the continuous layer of Fe3O4 is observed, it cannot represent the Fe3O4 layer in the reduced device geometry. Hence, we have monitored the 2-probe resistance of the device stack including the substrate instead of Fe3O4 layer alone which incorporates the high resistance tunnel junction region as well. Thus, it provides us more confidence in ruling out the extrinsic effects at the junction that may arise. Figure 3 is the plot of the 2-probe voltage of the device
Hence, it can be argued that the enhancement of spin signal is not due to the higher resistance of Fe$_3$O$_4$ at low temperatures, unlike the experimental conditions discussed by Wada et al. Using electroluminescence experiment. Since an additional barrier with 2–3 orders of higher resistance is used in our study, we do not observe any effect of the resistance enhancement of Fe$_3$O$_4$ layer. The trend in the enhancement of spin polarization observed by Wada and co-workers is gradual across the Verwey transition.

Following the preceding arguments, an anomalous increase in the spin accumulation voltage can be attributed purely to the spin sub-band modification across $T_V$ of Fe$_3$O$_4$ but not to the increase in resistance. Since the Verwey transition is associated with a structural modification, configuration of sub-bands of Fe$_3$O$_4$.

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**Figure 2.** Spin accumulation voltage as a function of temperature for devices of dimension 100 × 200 μm$^2$ on various systems. Plot of ΔV versus temperature of (a) Fe$_3$O$_4$/MgO/GaAs (b) Fe$_3$O$_4$/GaAs (c) Fe$_3$O$_4$/MgO/Si (d) Fe/MgO/GaAs devices. (e) Comparison of normalized ΔV for all the system. (f) Magnetization versus temperature for Fe$_3$O$_4$/MgO/GaAs, Fe$_3$O$_4$/GaAs and Fe$_3$O$_4$/MgO/Si thin film system showing Verwey transition (shown in shaded region) coinciding exactly the region of enhancement in ΔV as in (e).

**Figure 3.** Comparison of normalized value of local 2-probe voltages as a function of temperature for different devices on GaAs for 1 and 100 μA for devices on Si.
along with the tunnel barrier might influence the total spin accumulation, as \( \Delta V \) depends directly on the tunnel spin polarization of Fe\(_3\)O\(_4\)/MgO. But the similar magnitude of enhancement in spin accumulation is even present in the Schottky junction of Fe\(_3\)O\(_4\) on GaAs. Hence, the MgO electronic structure bands may not play any vital role in the observed enhanced values of \( \Delta V \) at lower temperatures.

Enhancement of \( \Delta V \) in Fe\(_3\)O\(_4\) based spin injection could be due to the enhancement of spin polarization in Fe\(_3\)O\(_4\) layer. Though Fe\(_3\)O\(_4\) is theoretically predicted to be half-metallic in nature \([10, 11]\), experimentally, it has always been a material whose spin-polarization is less than 100\% \([12, 28, 29]\) with values varying from 55 to 80\%. Fonin et al \([30]\) observed that the quality and the growth conditions of thin film highly influence the spin-polarization of Fe\(_3\)O\(_4\). For example, spin-polarization of (111) Fe\(_3\)O\(_4\) grown on (110) W is found to be \( \approx 80\% \), whereas (111) Fe\(_3\)O\(_4\) thin film grown on (11−20) Al\(_2\)O\(_3\) is only \( \approx 60\% \) spin-polarization and (001) Fe\(_3\)O\(_4\) grown on (001) MgO has spin-polarization of \( \approx 55\% \) \([31]\). In all the cases, the spin-polarization of Fe\(_3\)O\(_4\) is always higher than that of Fe, which is only 35\%. In addition, band structure calculations at lower temperatures also predict the half-metallicity of Fe\(_3\)O\(_4\) at \( T < T_V \) \([32]\). But experimental observations by Wang et al \([16]\) and Ziese et al \([15]\) show that the maximum spin-polarization observed at \( T_V \) using magnetoresistance measurements. Even in our case, the behavior of the spin accumulation voltage follows the same trend as discussed by Ziese et al \([15]\).

Hence, it is important to mention here that the present band structure calculations of Fe\(_3\)O\(_4\) predicts the half-metallicity even at lower temperature \( T < T_V \), i.e., invariance of spin-polarization below and above Verwey transition of Fe\(_3\)O\(_4\) \([10, 11]\). However our experimental results strongly suggest that the enhancement of spin accumulation in Fe\(_3\)O\(_4\) based devices is caused by an enhancement of spin-polarization across the Verwey transition. Hence, there is a need to revisit the band structure calculation of Fe\(_3\)O\(_4\) in order to solve the long standing puzzle of half-metallicity in Fe\(_3\)O\(_4\).

**Conclusions**

In conclusion, we have demonstrated all electrical spin injection and detection in various systems like GaAs and Si with the help of highly efficient spin injection from an oxide magnetic material Fe\(_3\)O\(_4\). Spin accumulation voltage measured in GaAs using 2 different spin injectors like Fe\(_3\)O\(_4\) and Fe is compared side-by-side, and the importance of a highly spin-polarized material like Fe\(_3\)O\(_4\) as a spin injector is justified. Also, in the absence of a tunnel barrier like MgO, spin injection and detection in Schottky junction of Fe\(_3\)O\(_4\)/GaAs is observed, in fact, with a higher magnitude of spin accumulation voltage. A huge enhancement in the spin accumulation voltage at \( T < 120 \) K is observed in the devices with Fe\(_3\)O\(_4\) as a spin injector, which is absent in devices with Fe films. From this it can be concluded that the \( T_V \) of Fe\(_3\)O\(_4\) film has a direct influence on the magnitude of spin accumulation and the resistance enhancement of Fe\(_3\)O\(_4\) film itself has no vital role in the observed enhancement of spin accumulation. Moreover, these results are consistent with the variation of spin polarization of Fe\(_3\)O\(_4\) with the temperature. Hence, from our study, it can be seen that the spin injection and detection into conventional semiconductors like GaAs and Si can be improved with the help of materials like Fe\(_3\)O\(_4\) with higher spin-polarization. Due to enhanced values of the spin accumulation, Fe\(_3\)O\(_4\) seems to be promising for the future semiconductor spintronics device applications.

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